A model for the production of regular fluorescent light from coherently driven atoms

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It has been shown in recent years that incoherent pumping through multiple atomic levels provides a mechanism for the production of highly anti-bunched light, and that as the number of incoherent steps is increased the light becomes increasingly regular. We show that in a resonance fluorescence situation, a multi-level atom may be multiply coherently driven so that the fluorescent light is highly anti-bunched. We show that as the number of coherently driven levels is increased, the spontaneous emissions may be made increasingly more regular. We present a systematic method for designing the level structure and driving required to produce highly anti-bunched light in this manner for an arbitrary even number of levels.

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I. INTRODUCTION

It is well known that resonance fluorescence from a two level atom is anti-bunched [1,2]. That is, consecutive fluorescent photons are less likely to arrive very close together (ie. to be bunched) than they are in a classical light beam of the same average intensity. The distribution of waiting-times between photo-detections for resonance fluorescence is therefore peaked further away from zero than for classical light. The reason for this effect is easily understood. When the atom spontaneously emits to produce a fluorescent photon it places itself in the ground state. It cannot emit again until the laser field driving the atom has increased the occupation probability of the excited state. The evolution of the excited state occupation probability amplitude for a driven two level atom (excluding spontaneous emission) is given in Fig.(1). It is the nature of this evolution (along with the spontaneous emission rate of the excited state) which determines the form of the waiting-time distribution.

We note that in recent years it has been shown that in laser models with more than two levels, it is possible to achieve sub-Poissonian statistics in the output [3]. In general sub-Poissonian statistics implies anti-bunching [4]. Ritsch *et al.* [5] have shown that highly sub-Poissonian light may be produced if the atomic state is recycled from the lower to the upper lasing level via a large number of incoherent pumping steps through intermediate levels. As the number of pumping steps tends to infinity the intensity fluctuations of the output light tends to zero. In this paper we will consider a single atom resonance fluorescence situation rather than a laser model. In this case the driving is completely coherent, and the anti-bunched light is produced by a single incoherent transition.

It has been shown previously that a coherently driven three level Raman laser gives a sub-Poissonian output [6]. Also, Schernthanner and Ritsch [7] have shown that detuning of the pump and laser light from the atomic transitions in a Raman (3-level) laser can lead to a value of the Mandel Q parameter close to -1, and hence a highly sub-Poissonian output. Ralph and Savage [8] have shown that a coherently driven 4-level laser will produce amplitude squeezed light. In this paper we treat atoms with an arbitrary, but even, number of levels.

We might call the limit of extreme anti-bunching *regular* light. That is, light in which the time between consecutive photon-detections is constant, or at least, the fluctuations in the temporal separation of consecutive photons is small compared to the average value of this separation. Clearly, if we can arrange the atomic evolution so that the occupation probability of an unstable state makes a sharp transition from zero to some nonzero value a well defined time after a spontaneous emission, then if we have a high spontaneous emission rate the result will be essentially regular fluorescence.

In the following section we show how we may design a coherently driven n level atomic system (where n is even) to produce an evolution in which the anti-bunching is increased over that for a two level system. The evolution may be made increasingly closer to a given desired evolution (for example that to produce regular light) as the number of levels is increased. In Section III we give an example of this procedure for the case of a four-level system. In Section IV we calculate the waiting-time distributions achieved for various n. We conclude in Section V.

II. TAILORING THE ATOMIC EVOLUTION

Excluding spontaneous emission, the equation of motion for the occupation probability amplitudes for a coherently driven n level atomic system may be written [9,10]

$$\frac{d}{dt}\begin{pmatrix} a_1 \\ \vdots \\ a_n \end{pmatrix} = -iH \begin{pmatrix} a_1 \\ \vdots \\ a_n \end{pmatrix}, \tag{1}$$

where a_j is the probability amplitude for state j, and H is an $n \times n$ Hermitian matrix, whose elements, h_{ij}, we will take to be real for simplicity. We are also using units

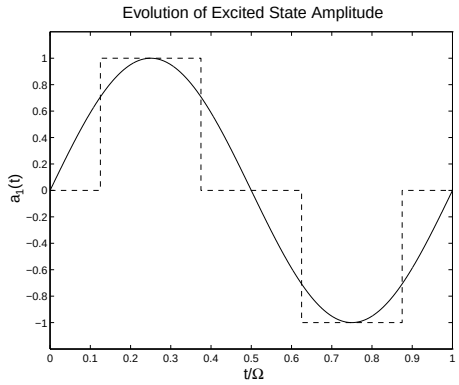

FIG. 1. Solid line: The evolution of the excited state probability amplitude of a driven two level atom with Rabi frequency Ω. Dashed line: An imaginary evolution that would produce light, the regularity of which would be limited only by the spontaneous decay rate of the excited state.

such that $\hbar = 1$. The matrix H, which is the Hamiltonian, is determined both by the laser fields driving the atomic levels, and the dipole matrix elements between the atomic levels. The magnitude of the off-diagonal element h_{ij} is given by the product of the strength of the laser field coupling the atomic levels i and j with the magnitude of the dipole matrix element connecting those levels. The phase of the off-diagonal element h_{ij} is determined by the the phase of the respective coupling laser field and the sign of the respective dipole element. The diagonal elements are determined by the detuning of the laser fields from the various atomic transitions being driven [11,12]. The general evolution (up until any spontaneous decay) is given by

$$\begin{pmatrix} a_1(t) \\ \vdots \\ a_n(t) \end{pmatrix} = \sum_{m=1}^{n} c_m e^{-i\lambda_m t}\mathbf{v}_m \ , \quad H\mathbf{v}_m = \lambda_m \mathbf{v}_m, \quad (2)$$

where the coefficients c_m are determined by the initial conditions chosen for the a_j. The $\mathbf{v}_m$ are the eigenvectors of H. We will write their components as $\mathbf{v}_m = (v_{1m}, \ldots, v_{nm})$, and assume them to be normalised so that $\mathbf{v}_m \cdot \mathbf{v}_l = \delta_{ml}$.

We now impose certain conditions on the laser interaction Hamiltonian H. We will show below how interaction matrices satisfying these conditions may be constructed. Once the interaction matrix has been constructed, this tells us the configuration of laser fields that is required to drive the atomic levels to produce the desired evolution. First of all let us assume that n is even, and that for every eigenvalue of H there is another eigenvalue which has the same magnitude but opposite sign, so that the eigenvalues come in pairs which sum to zero. Let us take the initial condition to be $a_n = 1$, and all the other amplitudes zero, so that the atom starts initially in the state n. Finally, let us assume that the n values $c_m v_{1m}$ also come in opposite pairs, such that $c_m v_{1m} - c_l v_{1l} = 0$ iff $\lambda_m - \lambda_l = 0$. The evolution of a_1 may now be written

$$a_1(t) = \sum_{m=1}^{n/2} 2ic_m v_{1m} \sin(\lambda_m t), \quad (3)$$

which is clearly a sine series for the evolution of a_1.

Before we show how to construct a matrix H with the above characteristics, let us turn to the question of regular photon emission. Let us assume that all the levels in our system have much lower spontaneous decay rates than level 1, and that this level decays via spontaneous emission to level n. For regular photon emission we therefore require that the evolution of the amplitude of level 1, given by the (finite) sine series above, have a sharp transition from zero to some non-zero value. An ideal evolution would be given, for example, by the dotted 'square' curve in Fig.(1). To approximate the ideal curve to a given level of accuracy we may choose the sine series in Eq.(3) to be the first n terms in the Fourier sine series for the desired 'square' evolution. The Fourier series in question is

$$f(t) = \sum_{m=1}^{\infty} \frac{2\sqrt{2}\mathrm{Im}[(1+i)i^m]}{(2m-1)\pi} \sin\left((2m-1)2\pi\Omega t\right), \quad (4)$$

where $1/\Omega$ is the period of f(t). Clearly the evolution will more closely follow the desired evolution as n is increased.

We should note here that we only require the evolution to be equal to the Fourier series up to a constant factor. This is because the overall scaling of the evolution is not important as far as the resulting waiting time distributions are concerned; we can cancel the effect of scaling the evolution curve by scaling the spontaneous emission rate of the unstable level with respect to Ω. This will be obvious when we write down the expression for the waiting time distribution in Section IV.

To construct the required matrix H we first write it in the form

$$H = TDT^t \quad (5)$$

where D is a diagonal matrix containing the eigenvalues of H, and T is a matrix whose columns are the normalised eigenvectors of H. Constructing H involves choosing the elements of D and T. We are clearly free to choose the eigenvalues of H, by choosing the diagonal elements of D. The second condition imposed above concerns the n values $c_m v_{1m}$. Now the c_m depend upon the initial conditions. Recall that we choose the initial conditions to be $a_1 = \ldots = a_{n-1} = 0, a_n = 1$. With this choice it is easily shown that $c_m = v_{nm}$. That is, the coefficient c_m is given by the last element of vector $\mathbf{v}_m$; the condition on the coefficients is a condition on the elements of the eigenvectors of H. In particular it is a condition on the first and last elements of the eigenvectors.

It is pertinent to note now that if the columns of a square matrix are orthonormal vectors, it follows that the rows are also orthonormal. To construct H we therefore require both to find a set of n orthonormal rows for

T, and satisfy the condition on the first and last elements of the vectors $\mathbf{v}_m$. The first elements of the $\mathbf{v}_m$ are the n elements of row one, and the last elements of the $\mathbf{v}_m$ are the n elements of row n. The condition we must satisfy is that the n values $v_{nm}v_{1m}$ must sum to zero in pairs, which clearly implies

$$\sum_{m=1}^{n} v_{nm}v_{1m} = 0. \tag{6}$$

This simply states that the dot product of row one with row n should vanish. The condition on the elements of the $\mathbf{v}_m$ is therefore consistent with the orthogonality of the rows.

The v_{1m} and v_{nm} may now be chosen so that the coefficients in the sine series for the evolution of a_1 match (up to an overall constant factor) the desired first n Fourier coefficients. Choosing the other $n-2$ rows of T is then a straight forward procedure in linear algebra. (For example the Gramm-Schmidt orthogonalisation procedure may be used [13]).

Note that there is clearly a degree of freedom in constructing the matrix H. If we take the matrix T to be real, then there are n^2 undetermined coefficients. Orthonormality of the rows imposes $n(n+1)/2$ conditions. Obtaining the desired Fourier coefficients imposes $(n-1)$ conditions (one less than n because the overall scaling of the coefficients is unimportant). This leaves $(n/2-1)(n-1)$ elements undetermined. These may therefore in general be chosen so as to simplify the form of H in order to simplify the corresponding physical system.

III. AN EXAMPLE FOR A FOUR-LEVEL SYSTEM

We now use the procedure described in the previous section. We construct a Hamiltonian for a four-level system that will produce evolution corresponding to the first two terms in the sine series given by Eq.(4).

First we choose the eigenvalues $-6\pi\Omega$, $-2\pi\Omega$, $2\pi\Omega$, $6\pi\Omega$, which give us respectively the diagonal elements of D. To satisfy the condition on the elements of T we first choose the top row of T to have elements all identical. As a result the bottom row of T must be chosen to have elements which vanish in pairs, the absolute value of each pair being proportional to the absolute value of a Fourier expansion coefficient. Thus we choose as the bottom row $(1,-3,3,1)/\sqrt{20}$. The other two rows are free to be any two further mutually orthonormal vectors. We decide to choose them such that three off diagonal elements of H are zero. The resulting T matrix is

$$T = \begin{pmatrix} 1/2 & 1/2 & 1/2 & 1/2 \\ -3/\sqrt{20} & -1/\sqrt{20} & 1/\sqrt{20} & 3/\sqrt{20} \\ 1/2 & -1/2 & -1/2 & 1/2 \\ 1/\sqrt{20} & -3/\sqrt{20} & 3/\sqrt{20} & -1/\sqrt{20} \end{pmatrix}. \tag{7}$$

The resulting Hamiltonian is given by

$$H = TDT^t = \frac{2\pi}{\sqrt{20}}\Omega \begin{pmatrix} 0 & -10 & 0 & 0 \\ -10 & 0 & -8 & 0 \\ 0 & -8 & 0 & 6 \\ 0 & 0 & 6 & 0 \end{pmatrix}. \tag{8}$$

Note that all the diagonal terms are zero. While the off-diagonal elements describe the coupling of the atomic energy levels via classical driving fields, the diagonal elements are determined by the detuning of the driving fields from the atomic transitions which they drive. The diagonal elements of the Hamiltonian are zero when all of the detunings are zero, so that the Hamiltonian we have constructed describes the situation where all the lasers are tuned to the transitions they drive. The value of Ω determines the strength of the laser fields, and may clearly be thought of as a generalised Rabi frequency. In Fig.(2) we give a diagrammatic representation of the four-level atomic system showing the coupling between the levels.

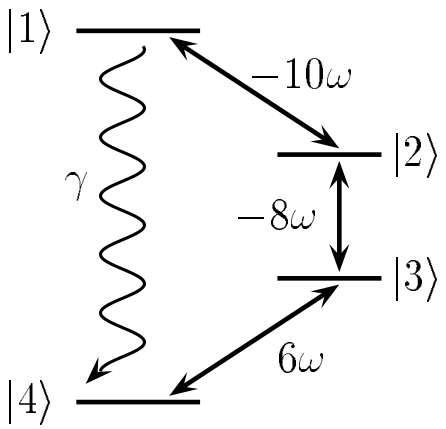

FIG. 2. A diagrammatic representation of the driven four-level atomic system designed to produce light with anti-bunching increased over a two level system. The elements of the Hamiltonian coupling the various levels are given as multiples of $\omega = 2\pi\Omega 20^{-1/2}$.

IV. WAITING-TIME DISTRIBUTIONS

We now derive the waiting time distribution from the evolution of the unstable state. Appealing to the Monte-Carlo Wave Function (or 'quantum trajectory') method for simulating the evolution of open quantum systems, the probability for a spontaneous photo-emission in time Δt is [14,2]

$$\gamma |a_1(t)|^2 \Delta t, \tag{9}$$

where γ is the spontaneous emission rate of level 1. First let $P(t)$ be the probability that there were no emissions in the interval $[0,t]$, given that there was an emission at time $t=0$. The probability that there is no emission in the interval $[0, t+\Delta t]$ is therefore

$$P(t+\Delta t) = P(t)(1-\gamma|a_1(t)|^2\Delta t). \tag{10}$$

Taking the limit as Δt tends to zero 0 we arrive at a differential equation for P(t), given by

$$\frac{dP(t)}{dt} = -\gamma|a_1(t)|^2 P(t), \tag{11}$$

the solution of which is

$$P(t) = e^{-\gamma\int_0^t |a_1(t')|^2\,dt'}. \tag{12}$$

The waiting-time distribution, $w(t)$, is then $P(t)$ multiplied by the rate of emission at time t. It is clear from Eq.(11) that this may be written

$$w(t) = -\frac{dP(t)}{dt} = \gamma|a_1(t')|^2 e^{-\gamma\int_0^t |a_1(t')|^2\,dt'}. \tag{13}$$

Clearly this equation will provide an analytic solution for $w(t)$ as long as the integral of $a_1(t)$ possesses an analytic solution. Naturally this is true for any truncated Fourier series. In Fig.(3) we plot the waiting-time distributions that would be generated by systems which produce an evolution matching the first n terms in the Fourier series given by Eq.(4), for various values of n. We calculate $w(t)$ by numerically evaluating Eq.(13), where $a(t)$ is given by Eq.(4). Clearly the waiting-time distribution is increasingly peaked for increasing n. Experimentally there is a great deal of freedom in choosing the generalised Rabi frequency Ω since it is determined by the intensity of the laser driving fields. For the plots in Fig.(3) we have used $\Omega = \gamma/100$ which is well within experimental limits.

Another quantity of interest is the second order correlation function of the fluorescent light, $G^2(\tau)$. This is the probability density for one photon to be emitted at time t and another photon to be emitted at time $t+\tau$, regardless of how many other photons where emitted in the intervening time interval $(t, t+\tau)$. The second order correlation function is given by $rQ(\tau)$, where $Q(\tau)$ is the probability that a photon is emitted at a time $t+\tau$, given that we have a photon emitted at time t, and r is the probability per unit time for a photo-emission. The latter is simply the average rate of photo-emission, which is

$$r = \left(\int_0^\infty w(t)t\;dt\right)^{-1}. \tag{14}$$

To calculate Q we may use $w(t)$, but we must sum over the probabilities for different numbers of photons to be emitted in the interval $(t, t+\tau)$ [15]. This is performed very neatly by the formula

$$Q(\tau) = w(\tau) + \int_0^\tau Q(t)w(\tau-t)\;dt. \tag{15}$$

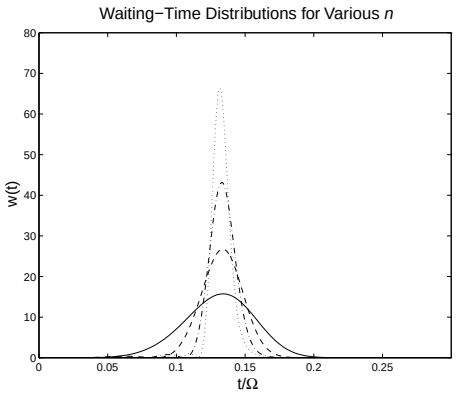

FIG. 3. Waiting-time distributions that would be generated by systems which produce an evolution matching the first n terms in the Fourier series given by Eq.(4), for various values of n. Solid line: $n = 2$. Dashed line: $n = 4$. Dash-dot line: $n = 8$. Dotted line: $n = 16$. The value of the spontaneous emission rate is $\gamma = 100\Omega$.

If we define $\tilde{Q}(z)$ and $\tilde{w}(z)$ as the Laplace transforms of $Q(\tau)$ and $w(\tau)$ respectively, then this equation may be expressed as

$$\tilde{Q}(z) = \frac{\tilde{w}(z)}{1-\tilde{w}(z)}. \tag{16}$$

For a given $w(t)$ we may therefore obtain the corresponding $Q(t)$, and therefore $G^2(t)$, by solving Eq.(15) using Laplace transforms. Alternatively the second order correlation function may be obtained by solving the master equation describing the atomic system, and using the quantum regression theorem [2]. For our purposes it is simplest to obtain $Q(\tau)$, and hence $G^2(\tau)$, by numerically integrating Eq.(15).

In Fig.(4) we plot the normalised second order correlation function, $g^2(\tau) = G^2(\tau)/r^2$, for various values of n. As we expect, this has a series of peaks and troughs which gradually even out with time. As the regularity of the photon emission increases, longer separation times, τ, are required before the correlation function becomes smoothed out.

We have considered in detail here only the ideal case in which spontaneous emission from atomic states other than level one can be ignored. However, the effects of spontaneous emission from the other atomic levels is easily estimated. In this scheme the evolution of the amplitude of level one, $a_1(t)$, and the ratio γ/Ω are chosen so that, excluding spontaneous emission from any other atomic level, level one will emit after a time delay of approximately $1/(8\Omega)$ (that is, approximately one eighth through the generalised Rabi cycle which has period $1/\Omega$). Clearly we may safely over estimate the amplitude of the other levels by taking them to be unity. The effect of another state with a decay rate of $\gamma_o = (8\Omega)/k$, where k is some dimensionless number, is therefore to

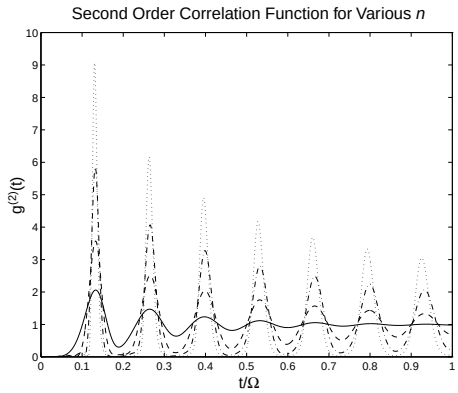

FIG. 4. The normalised second order correlation function that would be generated by systems which produce an evolution matching the first n terms in the Fourier series given by Eq.(4), for various values of n. Solid line: $n = 2$. Dashed line: $n = 4$. Dash-dot line: $n = 8$. Dotted line: $n = 16$. The value of the spontaneous emission rate is $\gamma = 100\Omega$.

interrupt the normal (regular) spontaneous emission sequence on average no more than once every k emissions. Taking $\gamma = 100\Omega$, which is the value we have used for the previous plots, then for a value of $\gamma_o = \gamma/10^4$, spontaneous emissions from this level will, on average, interrupt the desired sequence fewer than approximately once every 1000 emissions. Clearly in this case, and for larger ratios k, the waiting time distributions we have calculated will be a good approximation to the true distributions; any changes to the curves will be on the order of 1/1000 or less of the height of the main peak.

Clearly to implement our scheme experimentaly requires that, for a given atom, atomic levels can be found with very different Spontaneous emission rates. Hovever, this is not dificult, as spontaneous emmision rates for atomic levels are found to vary by many orders of magnitude. Observation of atomic shelving [16] (interruptions of the fluorescence from one 'bright' level by shelving to a meta-stable level) with single trapped ions has already been performed [17]. In these experiments the spontaneous emission rate of the meta-stable level is of the order of 10^{-8} of that of the bright level. An experimental realisation of our scheme, at least for the four level system which we have treated explicitly, therefore appears to be well within the limits of current technology.

V. CONCLUSION

We have shown that multiply coherently driven multilevel atomic systems may be designed to produce light which is more anti-bunched than for a two level system. We have given a procedure for deriving the driving required to produce highly anti-bunched light for a given number of atomic levels. This procedure requires that one of the atomic levels have a spontaneous emission rate which is much larger than the spontaneous emission rates of the other levels. We have also shown that as the number of levels is increased, the fluorescent light may be made increasingly regular.

We have treated explicitly the case of a four-level system, calculating a possible driving configuration to produce highly anti-bunched light.

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